TIME-RESOLVED LASER-INDUCED DYNAMICS OF EXCITED ALKALIS ON HELIUM CLUSTERS

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The dynamics of optically-excited monomeric and oligomeric alkali species formed on large liquid helium clusters ($n = 10^3 - 10^4$) have been probed in the picosecond time domain using the technique of time-correlated single photon counting. The guest atoms reside as surface species on the quantum cluster surface which serves as an excellent substrate for the study of fundamental chemical and physical dynamics. Wavelength-resolved fluorescence arising from wavelength-selective excitation yields information on the interaction of the non-degenerate P orbitals with the helium matrix which leads to the dynamical mixing of the $P_{3/2}, P_{1/2}$ states. Characterization of the intersystem crossing between the quartet and pre-dissociative doublet potential energy surfaces of Na_3 [a],is currently under way. The formation and decay lifetimes of the dissociation products will be determined.

[a] J. Higgins *et al.*, *Photoinduced Chemical Dynamics of High-Spin Alkali Trimers* Science, *273* 629-31, 1996.